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OPTICAL CONVERSION PROCESSES

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) The overall goal of this experimental program is to identify scalable techniques that efficiently convert existing high power UV lasers to lasers operating at longer wavelengths in the visible. Two non-linear optical conversion techniques that we have considered are: stimulated Raman and parametric conversion involving the KrF laser (248 nm). The objective of this contract is to suggest likely acceptor atoms for each technique which will thereby allow us to evaluate some of the key technical		

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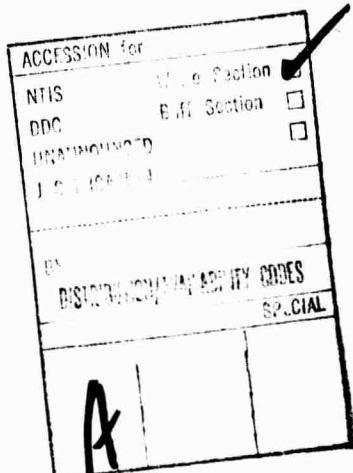
issues involved. These include: the production of receptor candidates in the gas phase (typically refractory metals), the volumetric removal of the lower laser level in the stimulated Raman approach to prevent "bottle necking" and allow recycling of the atoms during the laser pulse, and the consideration of overall system efficiency and scalability to high power.

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1. INTRODUCTION

The overall goal of this combined experimental and theoretical program is to successfully and efficiently convert using scalable techniques the output of a high power KrF laser into longer wavelengths so as to vastly improve its propagation characteristics.

Since the first reported lasing of an inert gas halogen laser, a number of similar systems have demonstrated lasing characteristics. Operating at various wavelengths, with different efficiencies, a major class of electronic transition lasers came into existence. Recently, analogous mercury halide compounds showing similar formation kinetics have been shown to lase in the visible,^(1, 2) albeit in high temperature ($\sim 275^{\circ}\text{C}$) cells (see Figure 1). However, the most efficient laser reported to date in this group is the KrF laser operating at 248 nm. It has also produced the highest energy outputs reported utilizing e-beam pumping and e-beam controlled discharge pumping and has a demonstrated capability for being scaled to high average power. In certain applications, especially those requiring transmission through the atmosphere, its short wavelength severely limits its usefulness. The limitation in atmospheric propagation at short wavelengths arises due to absorption by ozone in the atmosphere and to Rayleigh scattering which increases as λ^{-4} as the wavelength gets shorter. Ozone absorption is severe for wavelengths $\lesssim 3000 \text{ \AA}$.

Figure 2 shows vertical transmission from a height of 3 km as a function of wavelength. Also plotted are quantum efficiency of conversion from KrF wavelengths and the total percentage transmission of converted

(1) Parks, J.H., private communication.

(2) Parks, J.H., private communication.

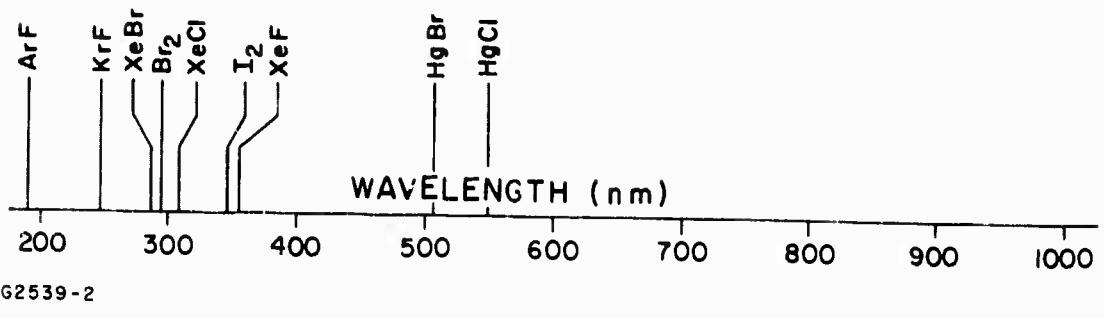


Figure 1 Demonstrated Potentially Scalable Electronic Transition Lasers

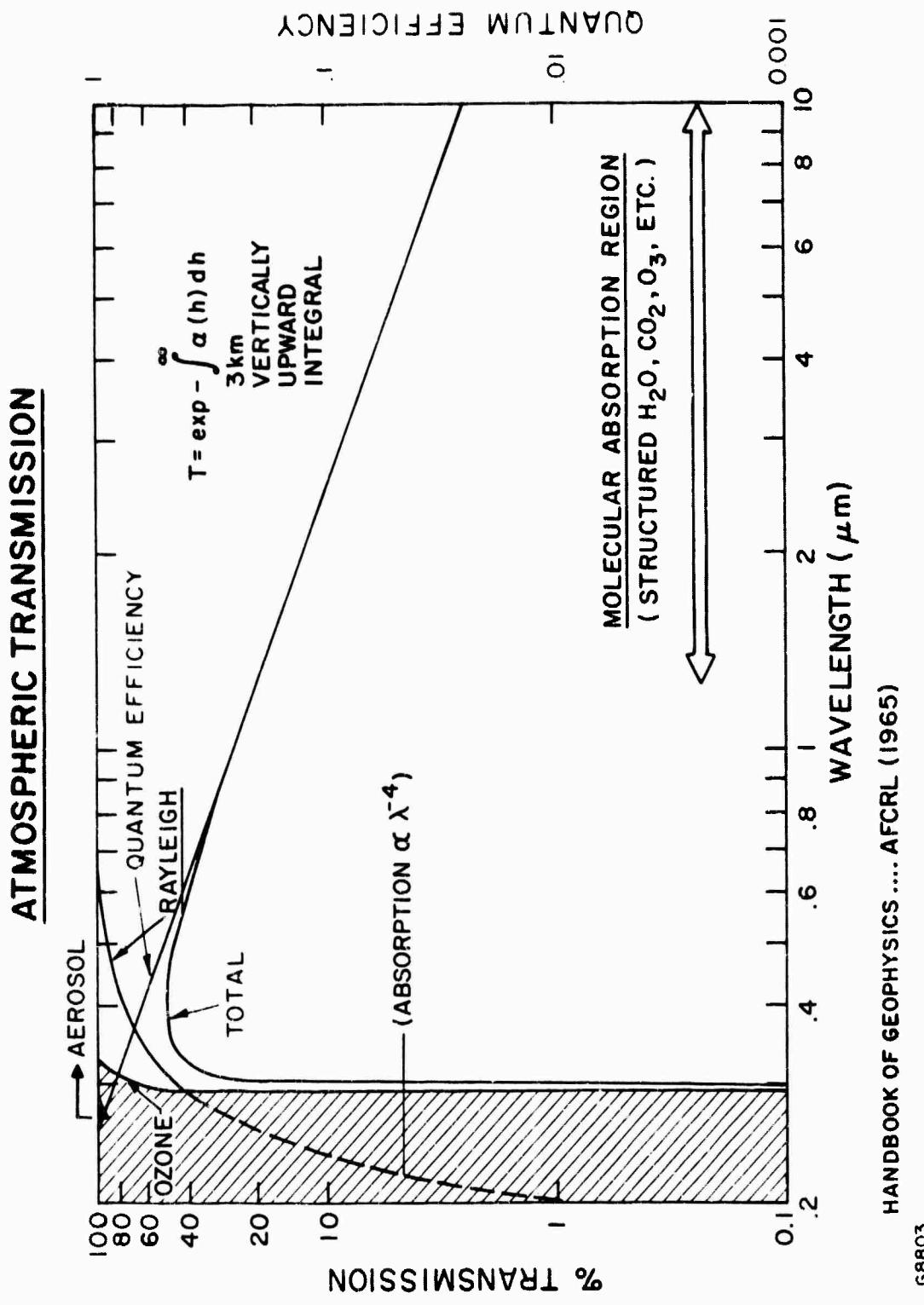


Figure 2 Propagation of Converted KrF Photons

KrF radiation. From the figure, it is apparent that to efficiently utilize KrF laser radiation, its conversion wavelength should be between 340 and 400 nm to maximize its atmospheric transmission with minimal loss from quantum yield considerations. Xenon fluoride lasers, while possessing a more attractive wavelength for propagation, have not yet demonstrated the combined efficiency and energy density comparable to KrF. Any optical conversion scheme for altering the wavelength of KrF laser radiation to the 340 to 400 nm wavelength range could have higher overall efficiency than the XeF laser if the photon conversion efficiency is $\gtrsim 40\%$. Such efficiency for conversion is a reasonable goal for the program we are discussing here. For supporting evidence, one can look over the past year at a number of milestones that have been reported relevant to the optical conversion of UV excimer lasers. With regard to overall conversion efficiency, XeF laser output has been converted, at near unit photon conversion efficiency, using barium vapor.⁽³⁾ Also KrF conversion to a series of UV-visible lines due to 6 Stokes and 2 anti-Stokes transitions in high pressure molecular hydrogen was reported showing good overall conversion efficiency.⁽⁴⁾ In view of the above, it seems reasonable and important to develop scalable techniques that could efficiently convert KrF laser output to longer wavelengths. Two non-linear optical conversion techniques that we have considered to achieve this goal are the stimulated Raman and the parametric conversion processes.

For the stimulated Raman process, phenomenologically, the acceptor atom can be thought of as absorbing the incident KrF photon thereby making the transition to an excited virtual state and then, with the emission of a Raman photon at longer wavelengths, proceeding to a level near the ground

(3) Djeu, N. and Burnham, R., Park City, Utah (Feb. 1977).

(4) Loree, T. R., Sze, R. C. and Barker, D. L., Park City, Utah (Feb. 1977).

(initial) state. Through collisions with an efficient quenching gas, it can return to the initial state for subsequent re-excitation by the KrF laser field, i.e., exhibit high efficiency by recycling the metal atoms. The Raman process is enhanced when the virtual state is close to a real state.

Another method of "down conversion" to lower energy, longer wavelength photons applicable to UV laser light is parametric down conversion. In this process, conversion is achieved by the utilization of non-linear properties of the medium (the acceptor atom or molecules). Here an atom in state 0 upon exposure to KrF laser light of frequency ν_1 goes to a virtual state 1 and re-emits three photons of frequencies ν_2 , ν_3 and ν_4 such that $\nu_1 = \nu_2 + \nu_3 + \nu_4$. At the end of this process, the atom returns to its initial state entirely by optical transitions. Once again, if the various atomic transitions (ν_1 , ν_2 , ν_3 , and ν_4) in the acceptor are allowed and the dipole moments are large, near resonance effects enhance the overall process such that efficient down-conversion should be likely.

At AERL during the current reporting period, theoretical and experimental research have been carried out on potentially efficient scalable schemes for converting KrF photons to longer wavelengths. By theoretical calculations, we have identified a number of promising candidates to convert the KrF laser radiation to longer wavelengths (see Table 1). Experimentally we have investigated metal atom production techniques to produce acceptor candidates for stimulated Raman scattering experiments. In particular, we have produced metal atoms (Pt, Pb) in the gas phase at room temperature using flash photolysis techniques and discharge dissociation. The results of these combined theoretical and experimental efforts are summarized in the following sections of this report.

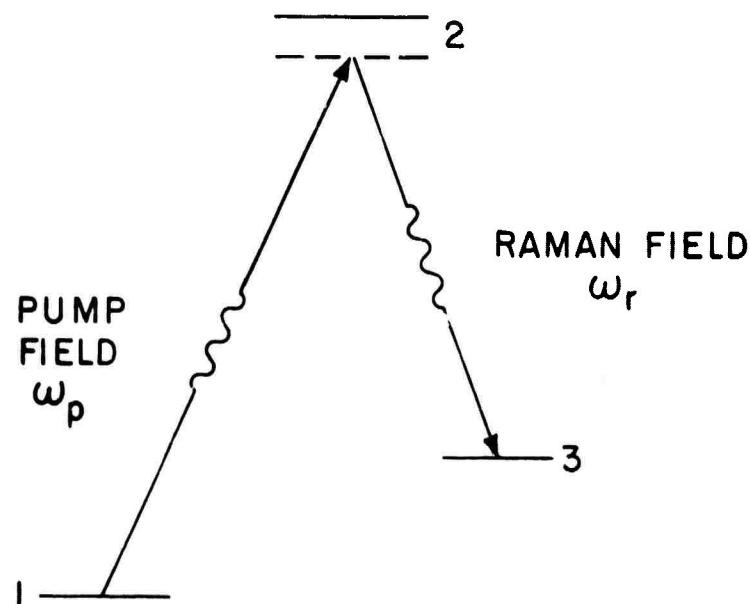
TABLE 1. POTENTIAL RAMAN DOWN CONVERSION CANDIDATES
AND THEIR OUTPUT WAVELENGTHS

Candidate	Output Wavelength λ , nm
Iron	300, 304
Calcium	544
Palladium	332
Platinum	332
Lead	309
Hydrogen	277, 313, 304

II THEORY

The stimulated Raman process is one of the earliest discovered non-linear optical processes since the advent of high power lasers. A schematic sketch of the Raman process is shown in Figure 3. Energy Levels 1 and 3 of the atomic or molecular medium are assumed to be connected through an allowed intermediate level 2. If light of energy close to the energy difference between levels 1 and 2 travels in the medium, one observes signals at the Raman frequency whose energy differs from that of the incident light by the energy difference between levels 1 and 3. Phenomenologically, the atom can be thought of as absorbing the incident photon, making a virtual transition to an excited state and then returning to the level 3 by emitting a Raman photon. Energy is conserved in the Raman process, the difference in energy between the incident (pump) photon and Raman photon being taken up (see Figure 3) or given by the atom. In the case where the atom gains energy, the process is called a Stokes process, while in the anti-Stokes process, the atom gives up some energy. The Raman process can be stimulated with an electromagnetic field at the Raman frequency. The Raman process is also enhanced considerably when the virtual state coincides or lies close to a real allowed state. When the virtual state coincides exactly with a real allowed state, both direct optical pumping and resonance Raman scattering will take place. At low intensities of pump power, optical pumping and lasing (stepwise single quantum transitions) dominates, while at high intensities, the Raman

— REAL STATE
- - - VIRTUAL STATE



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Figure 3 Schematic of Raman Process

scattering which is a double quantum transition is predominant. In the latter case, the output pulse spectrum will closely follow that of the pump spectrum.

Theoretical efforts under the present contract were concentrated in obtaining the stimulated Raman emission cross sections for the various atomic systems. A knowledge of the maximum density of metal atoms that one could produce and the Raman cross section enables one to calculate the maximum single pass gain of the Raman laser. A theoretical estimate of the gain for the various candidates will help the experimental program choose the best species.

The stimulated Raman emission cross section, σ_{SRE} , is related to the third order nonlinear Raman susceptibility, $\chi_r^{(3)}$, by the formula

$$\sigma_{SRE} = i \frac{64\pi^3}{\lambda_s^3 c} \chi_r^{(3)} \tilde{I}_p, \quad (1)$$

where λ_s is the wavelength of the Stokes photon and \tilde{I}_p is the pump laser flux in ergs/cm²/sec. The calculation of the nonlinear susceptibility, $\chi_r^{(3)}$, is usually a tedious exercise. It is given by the expression,

$$\chi_r^{(3)} = \frac{-i}{4 \hbar^3 \Gamma_{ca}} \sum_{a,c} (\rho_{aa} - \rho_{cc}) \left| \sum_b \left(-\frac{\langle c | \epsilon_s^* \cdot \vec{Q} | b \rangle \langle b | \epsilon_p \cdot \vec{Q} | a \rangle}{\Omega_{ba} - \omega_p} + \frac{\langle c | \epsilon_p \cdot \vec{Q} | b \rangle \langle b | \epsilon_s^* \cdot \vec{Q} | a \rangle}{\Omega_{ba} + \omega_s} \right) \right|^2 \quad (2)$$

where ρ_{aa} and ρ_{cc} are the fractional populations of initial and final states of the atom, \vec{Q} is the dipole operator ($= e \vec{r}$), $\Omega_{ba} = (E_b - E_a)/\hbar$, ω_p and ω_s are the pump and Stokes frequencies respectively and Γ_{ca} is the reciprocal of the dephasing time between levels c and a. ϵ_p and ϵ_s are the spherical unit polarization vectors of the pump and Stokes fields respectively. Yuratich

and Hanna⁽⁵⁾ have used Racah algebra to sum over the intermediate state M values and write the nonlinear susceptibility in a form that can be factored into two parts, in one of which the relation between the fields is explicitly displayed; the other contains the physics of the atom in the form of reduced matrix elements (which can be related to oscillator strengths) and 6j symbols. In terms of the total angular moment, J_ℓ , of the initial, final, and intermediate states and the reduced matrix elements, the Raman susceptibility $\chi_r^{(3)}$ can be written as

$$\chi_r^{(3)} = \frac{-i}{4\hbar^3 \Gamma_{ca}} \left(\frac{\rho_{aa}}{2J_1 + 1} - \frac{\rho_{cc}}{2J_3 + 1} \right) \sum_{K=0}^2 \Theta^{(K)} \left| \sum_{\gamma_2 J_2} \left\{ \begin{smallmatrix} J_3 & K & J_1 \\ 1 & J_2 & 1 \end{smallmatrix} \right\} \langle \gamma_1 J_1 | | Q | | \gamma_2 J_2 \rangle \langle \gamma_2 J_2 | | Q | | \gamma_3 J_3 \rangle \right|^2 \quad (3)$$

where $\Theta^{(K)}$ contains all the information about the angular dependence between pump and Raman fields and $\langle \gamma_a J_a | | Q | | \gamma_b J_b \rangle$ is the reduced matrix element. In a near-resonant Raman process, the factor $(\Omega_{\gamma_2 J_2 \gamma_1 J_1} - \omega_p)$ is very small for a particular state $\gamma_2 J_2$ and the summation in (3) is usually dominated by a single term. We can also assume that $\rho_{aa} = 1$ and $\rho_{cc} = 0$ at the beginning of the pulse. Combining (1) and (3), the stimulated resonant Raman cross section can be written as

$$\sigma_{SRE} \approx \frac{9}{8\pi^2} \frac{r_e^2}{\hbar c^2} \left(\frac{\nu_s}{\nu_{23}} \right) \frac{f_{12} (gf)_{32}}{\nu_{21} (\Delta\nu)^2} \frac{\tilde{l}_p}{\gamma_\ell} \times \sum_{K=0}^2 \left\{ \begin{smallmatrix} J_3 & K & J_1 \\ 1 & J_2 & 1 \end{smallmatrix} \right\}^2 \Theta^{(K)} \quad (4)$$

(5) Yuratich, M.A. and Hanna, D.C., J. Phys. B: Atom Molec. Phys. 9, 729 (1976).

In the above, r_e is the classical electron radius ($= 2.818 \times 10^{-13}$ cm), f_{ab} is the absorption oscillator strength from a to b, g is the degeneracy of the lower state and ν_{ab} is the transition frequency from a to b in cm^{-1} . Γ_{ca} in (3) has been replaced by $2\pi c \gamma_\ell$ where γ_ℓ is the laser line width in cm^{-1} , since at these frequencies, the laser line width is usually larger than any line width of the atom. $\Delta\nu$ ($= \nu_{21} - \nu_p$) is the amount of detuning of the laser line from the resonance line.

The total gain, $g_o L$, for a single pass system is equal to $N \sigma_{SRE} L$, where N is the density of atoms and L is the active length of Raman medium. For a diffraction limited beam, the maximum value of the intensity-length product is equal to P/λ_p where P is the pump laser power and λ_p is the pump wavelength. Thus, $g_o L$ can be written as,

$$g_o L = \Lambda N P / \gamma_\ell \quad (5)$$

where

$$\Lambda = 9.55 \times 10^{-14} \left(\frac{\nu_s}{\nu_{23}} \right) \frac{f_{12}(gf)_{32}}{(\Delta\nu)^2} \sum_{K=0}^2 \begin{Bmatrix} J_3 & K & J_1 \\ 1 & J_2 & 1 \end{Bmatrix}^2 \Theta^{(K)} \text{cm}^2/\text{W}$$

and P is in watts. The quantity Λ is an atomic parameter for a given pump wavelength and given pump and Raman field polarizations. In what follows, we shall assume for convenience, that both pump and Raman fields are linearly polarized and are traveling in the same direction.

In Figure 4, a map of constant $g_o L$ is plotted in the coordinates of Λ and N . Each atomic system is represented on this map by a horizontal line, whose position depends upon the density of the medium that may be produced in the gas phase and extent represents the degree of uncertainty in experimentally achievable atom production. The lines of constant $g_o L$ have been plotted on the assumption that P/γ_ℓ of the pump laser equals 10^5 W/cm.

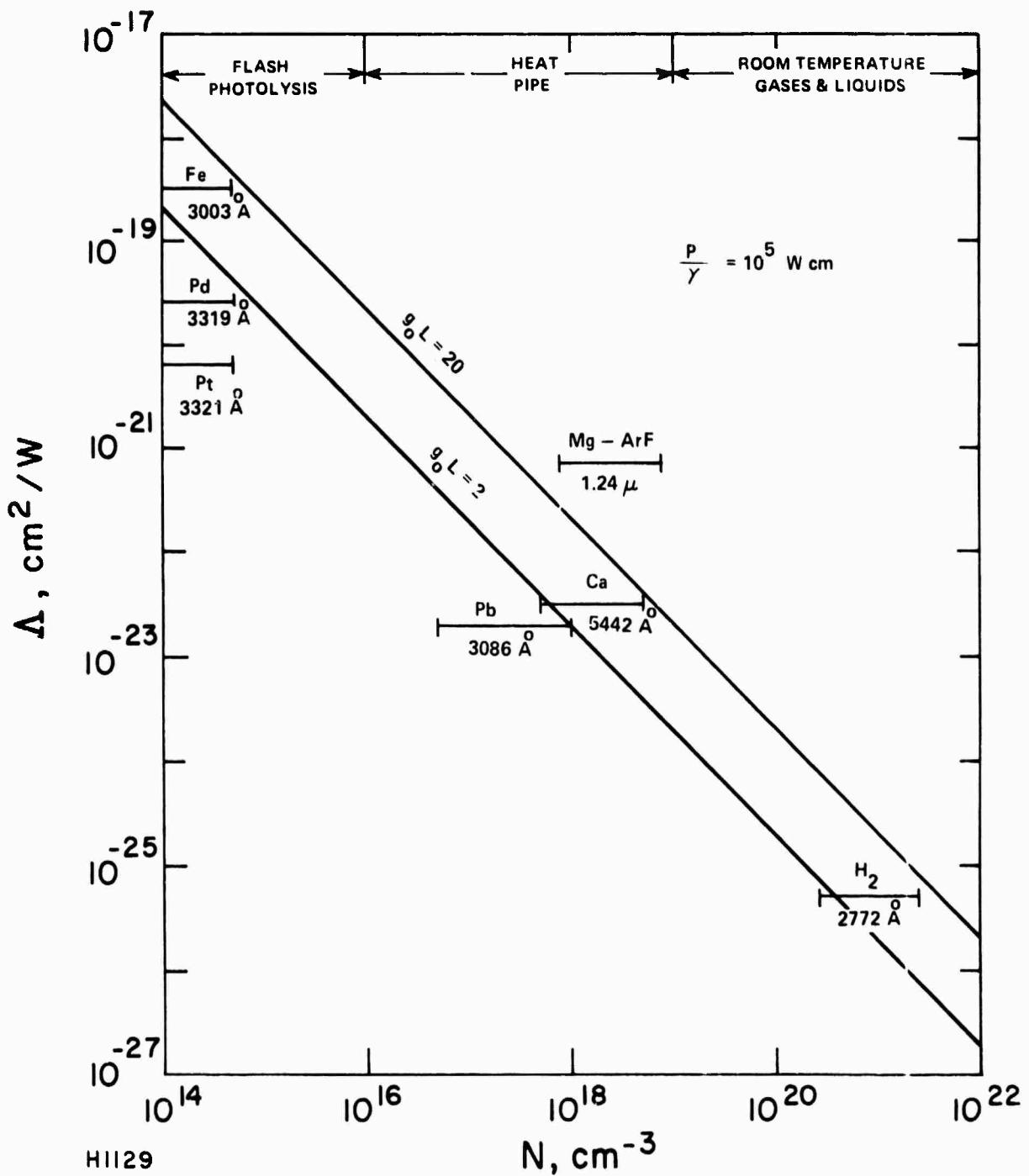


Figure 4 Anticipated Stimulated Raman Gain of Various Acceptor Candidates

For example, the specifications for a commercial Tachisto KrF laser are near 5 MW output over a bandwidth of $\sim 100 \text{ cm}^{-1}$, P/γ_ℓ is then half of this number. For a high power laser system, P/γ_ℓ is expected to be greater than 10^5 W/cm . For larger P/γ_ℓ , the lines of constant $g_o L$ shift towards the left. High gain systems fall to the right of the $g_o L \approx 20$ line. The map can be conveniently divided into three operating density regions based on the production of atoms or molecules. Low density atoms in the gas phase can be produced by photodissociation of organometallic compounds. Medium densities (10^{16} to 10^{19} cm^{-3}) are conveniently produced in a heat pipe or oven, while high densities usually correspond to room temperature liquids or high pressure gases. The division is not very rigid as low density gases can be produced by variety of techniques. A number of potential atomic systems are displayed on the map as well as molecular hydrogen.

For KrF down conversion, the most promising conversion systems, in terms of ease of demonstration of principle, are molecular hydrogen, atomic iron in the gas phase, and calcium vapor in a heat pipe. Although platinum does not show as large a projected high gain as does atomic iron, it has the advantage of potentially shifting the KrF output to propagating wavelengths. One aspect many of these systems have in common is the effect of direct optical pumping as a competing process to near resonant or resonant Raman processes. It was useful therefore to consider these effects using iron as an example.

The resonance absorption cross section σ_{abs} is given by

$$\sigma_{\text{abs}} = \pi r_e \frac{f}{\Delta\nu}$$

where f is the absorption oscillator strength and $\Delta\nu$ is the width of line.

The rate of pumping the upper level is given by $\sigma\phi_\nu$ where ϕ_ν is the photon

flux in a bandwidth of $\Delta\nu$ centered around ν . If ϕ is the total laser flux and γ_ℓ is its width,

$$\phi_\nu = \phi \frac{\Delta\nu}{\gamma_\ell}$$

$$\sigma_{\text{abs}} \phi_\nu = \frac{\pi r_e f}{\gamma_\ell} \phi$$

$$= \frac{\pi r_e f}{\gamma_\ell} \frac{I_L}{h\nu}$$

For the Fe transition from 0 to 40257 cm^{-1} , the rate of pumping the upper state is $3.8 \times 10^5 I_L / \gamma_\ell \text{ sec}^{-1}$ where I_L is in W/cm^2 and γ_ℓ is in cm^{-1} . Taking I_L to be 10^6 W/cm^2 and γ_ℓ to be 100 cm^{-1} , the pumping rate is larger than 10^9 sec^{-1} . This rate is much larger than the spontaneous emission rate from the upper state, and so we can expect the ground state and upper state populations to come into equilibrium within 1 nsec, which is much shorter than the KrF laser pulse ($\sim 20 \text{ nsec}$). The normal 2-level laser gain is given by

$$g_o = \frac{\lambda^2}{8\pi} \frac{A \Delta N}{\Delta\nu}, \text{ cm}^{-1}$$

where ΔN is the inversion density. If the pump laser intensity is not very high, the lasing transition is broadened only by collisions and doppler effects. Taking $N = 10^{14} \text{ cm}^{-3}$, $\Delta N = N/2$ and $L = 30 \text{ cm}$, we find that

$$(g_o L)_{\text{2-level}} \approx 750 \text{ while}$$

$$(g_o L)_{\text{Raman}} \approx 3 \text{ for this case.}$$

It thus appears that the gain due to direct pumping of the upper state, whenever the KrF laser line overlaps the resonance transition will be much larger

than the Raman gain. The converse will be true, if the KrF laser line is narrow and its frequency spectrum does not overlap the transition to the upper state. These conclusions are not general however because the line widths depend on pressure and temperature of the medium as well as pump laser intensity. One should therefore calculate for each system which gain would be dominant. In the case of the Fe and Pt, an un-narrowed KrF laser line will overlap the transition to the upper state and so in these two cases, one may expect direct optical pumping and lasing to be a dominant mechanism for down conversion.

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III. EXPERIMENTS

Experimental efforts during this reporting period have been concentrated on metal atom production and KrF laser procurement for use as a source for conversion and stimulated Raman experiments. We will briefly describe the significant milestones accomplished in each of these areas.

In that most of the identified acceptor candidates for efficient conversion were refractory metals, our initial program involved quantified measurements of the achievable densities we could produce in a scalable way in the gas phase. Two techniques were developed and measurements made which indicate that platinum atom densities in excess of 10^{13} atoms/cm³ could be produced in low pressure discharge devices and greater than 10^{11} atoms/cm³ of metastable lead atoms in our existing flash photolysis device developed under prior ARPA contracts. (See Table 2.) As an example, for platinum these densities correspond to operating a platinum thermal source at temperatures in excess of 2000°K.

TABLE 2. METAL ATOM DENSITIES DEMONSTRATED

Candidate	Technique	
	Flash Photolysis	Discharge
Platinum	Inconclusive	$> 5^{13}$ atoms/cc
Lead (3P_0)	$> 1^{13}$ atoms/cc	
(3P_1)	$> 5^{11}$ atoms/cc	

These two experimental techniques, flash photolysis and glow discharge dissociation, are described schematically in Figures 5 and 6 respectively.

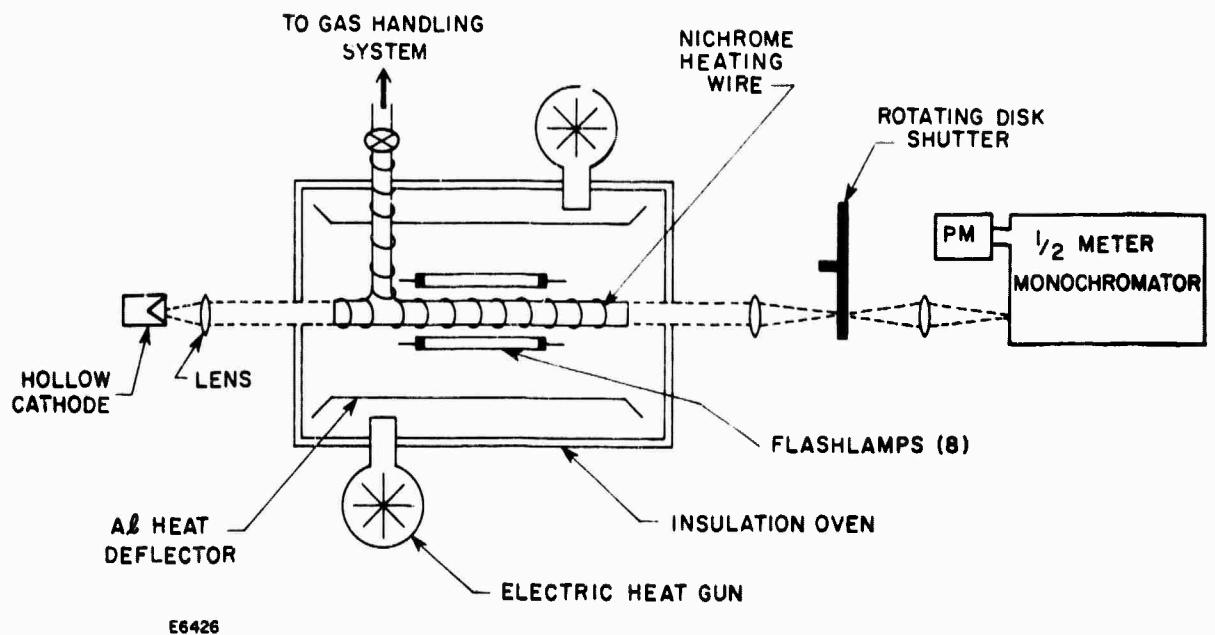
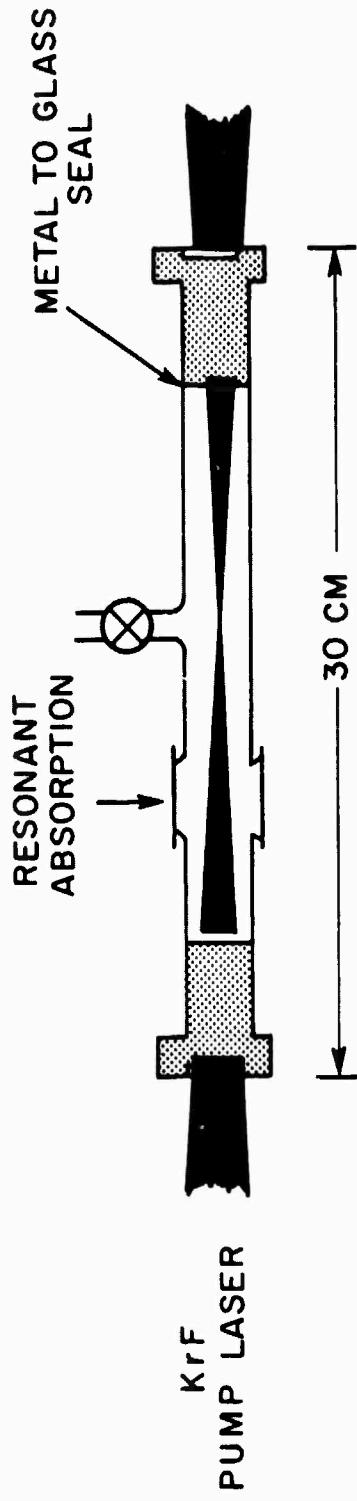
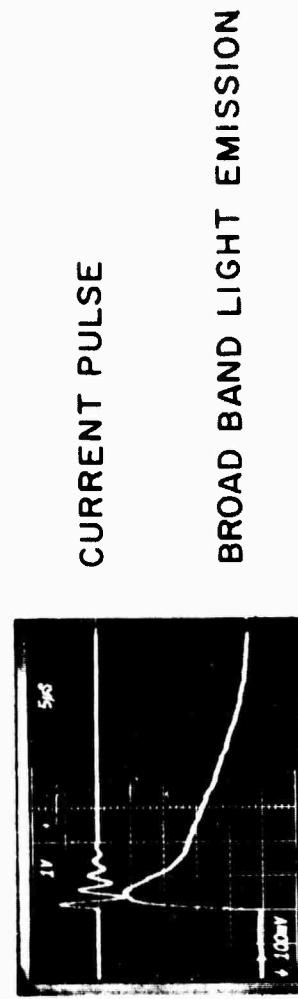


Figure 5 Diagram of Flash Photolysis Apparatus



50 TORR NEON + 0.1 TORR ORGANO-METALLIC



H1933

Figure 6 Discharge Cell Characteristics

By using a number of hollow cathode sources to perform absorption measurements, we were able to show qualitatively that we could produce atomic platinum by either technique (see Figure 7). The flash technique, however, seemed to also produce an absorbing fragment which persisted for several seconds (see Figure 8). This may introduce complications for later lasing experiments. These experiments clearly demonstrated that needed metal atom densities as described in our proposal can be produced for use as acceptor candidates to perform optical conversion lasing experiments.

To provide a pump laser to perform these experiments, we have taken delivery of a dye laser (Phase-R Corp, New Durham, N.H.) which puts out ~ 1/2 J at 5000 Å light in 500 nsec using Coumarin 504 dye (Exciton Corp, Dayton, Ohio). This laser is partial delivery of a doubled dye system which will utilize a potassium pentaborate doubling crystal to provide tunable output in the ultraviolet for use as a pump to perform conversion experiments.

(PLATINUM ACETYL ACETONATE, 10 TORR NEON, 4 J DISCHARGE)

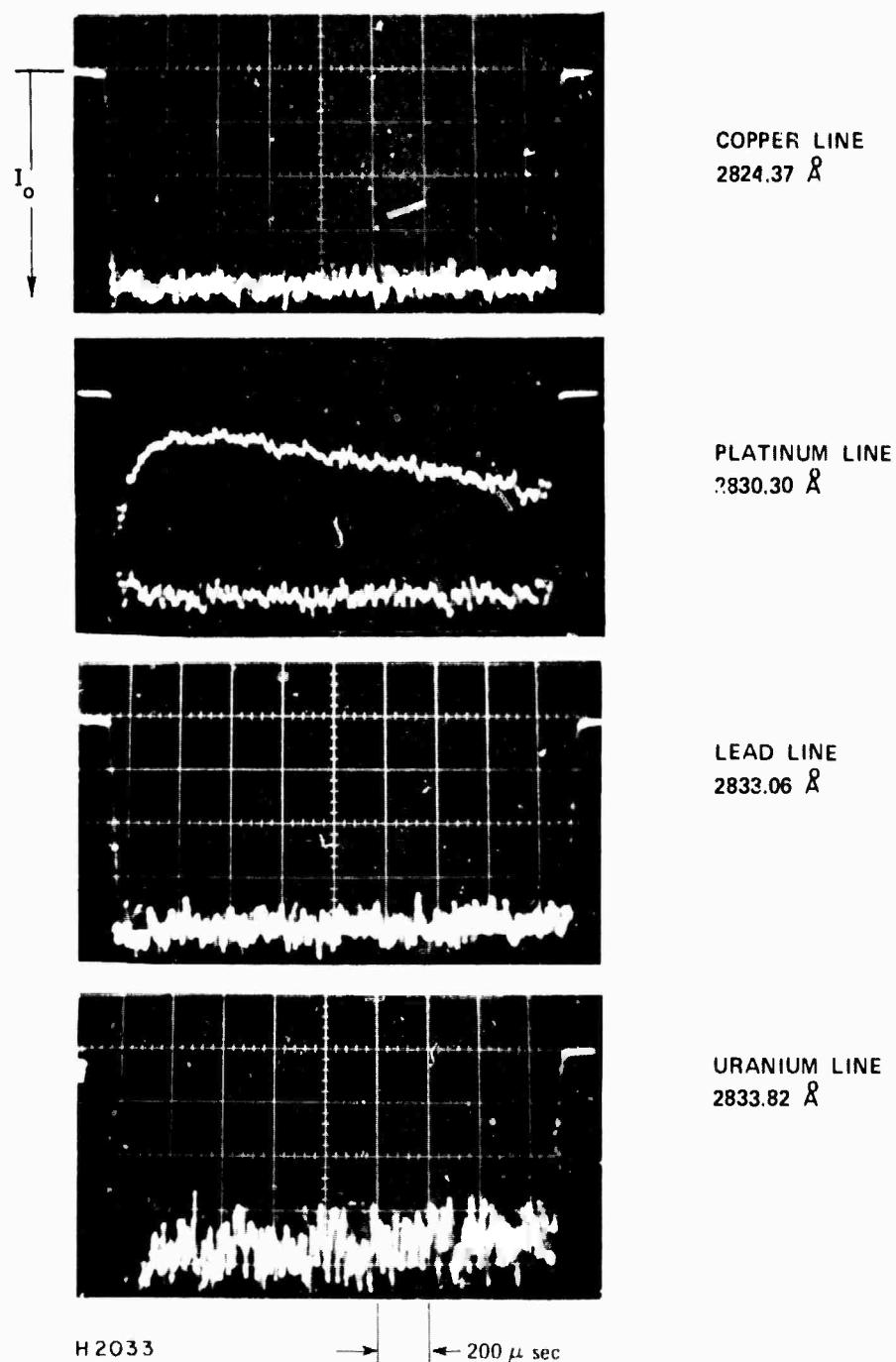
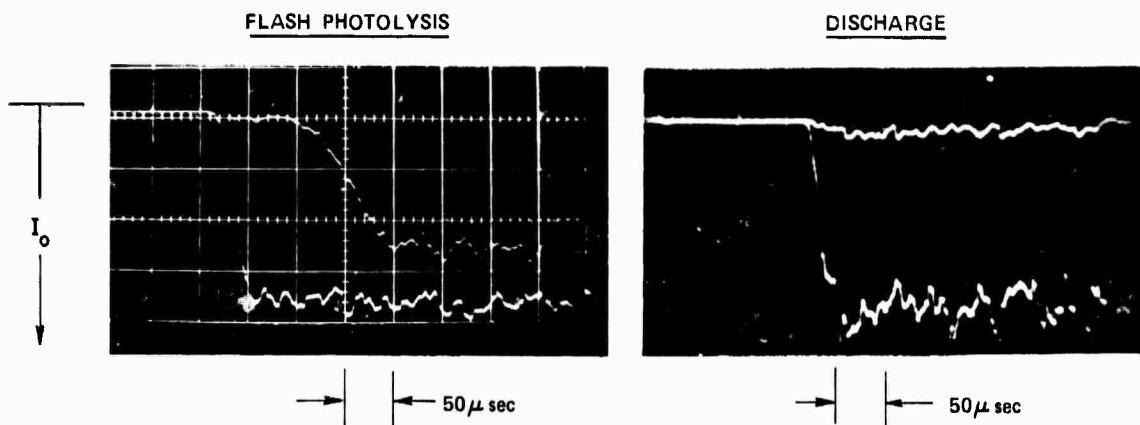


Figure 7 Platinum Production: Atomic Absorption Measurements



CYCLOPENTADIENYL TRIMETHYL PLATINUM		—	COMPOUND	—	CYCLOPENTADIENYL TRIMETHYL PLATINUM
ARGON		—	BUFFER	—	NEON
400 J		—	ENERGY	—	4 J
20 cm		—	PATH LENGTH	—	20 cm
$9.4 \times 10^{-13} \text{ cm}^2$		—	σ	—	$9.4 \times 10^{-13} \text{ cm}^2$
H 2034	3064.71 Å	—	λ	—	3064.71 Å

Figure 8 Atomic Platinum Production: Flash vs Discharge

IV. SUMMARY

To meet the long range goal of ARPA of producing high power laser radiation in the visible wavelengths, theoretical and experimental research have been carried out on potentially efficient scalable schemes for converting KrF photons to longer wavelengths. Experimental investigation of metal atom production techniques to produce acceptor candidates for stimulated Raman scattering experiments have been carried out. In particular, we have produced metal atoms (Pt, Pb) in the gas phase at room temperature using flash photolysis technique and discharge dissociation of organometallic compounds. By means of theoretical calculations, a number of promising candidates for the Raman conversion of KrF laser radiation have been identified.

Actual demonstration of down conversion will be carried out in the coming months. Theoretical research will be carried out on parametric down conversion, emphasis being placed on the aspects of phase matching, calculation of the nonlinear susceptibility and conversion efficiency.

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